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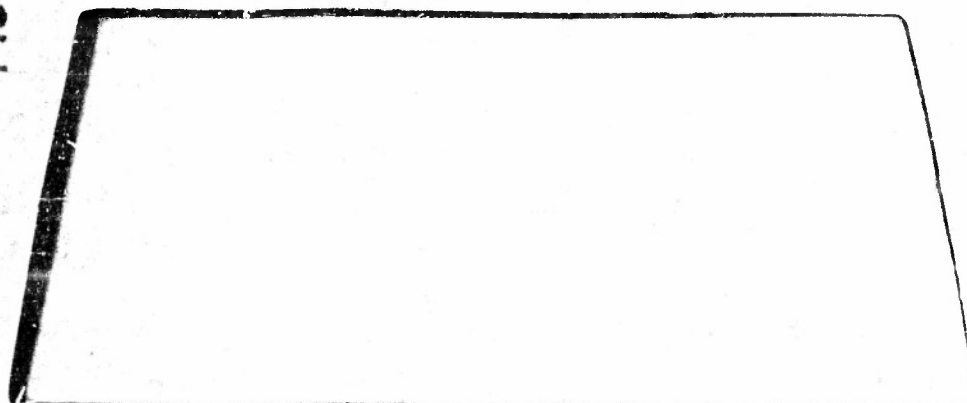
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Syracuse University

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A MOMENT OF SINGULARITY ANALYSIS
OF VIBRATION SPECTRA

Melvin Lax,^{*} and Joel L. Lebowitz[†]

^{*} Principal Investigator

[†] Not employed on project NR 017-419

Abstract

The frequency distribution of a crystal is approximated by combining Van Hove's determination of its analytical nature and Montroll's method of moments. The function $G(\omega^2)$ is represented by an expression with the correct behavior at the singularities and at the maximum and minimum frequencies. The behavior between singular points is adjusted smoothly by leaving n undetermined parameters. These parameters are then fixed by using the correct first n moments. As a test, this procedure was applied to the two dimensional square lattice with nearest and next nearest neighbor interactions, solved exactly for a particular case by Montroll. The approximated distribution function had the right form at the end points, contained terms of the appropriate logarithmic form and a jump function (with known coefficients). It also included Legendre polynomials with unknown coefficients, which were determined by the moments. The difference between the exact and approximate distribution functions was a few per cent using only the zeroth moment (normalization). Using higher moments produced a gradual increase in accuracy.

Introduction

The normal modes of a lattice are eigen-vectors of a matrix $M_{ij}^{\alpha\beta}$, denoted simply by M , where i, j are indices of the cell, and α, β are indices of particles in a cell. Using the translational symmetry of the lattice, the problem is customarily reduced to the diagonalization of a matrix $M^{\alpha\beta}(k)$, denoted briefly by $M(k)$, where k is the propagation constant of the wave, and the index α (or β) takes on ℓZ different values, where ℓ is the dimension of the space and Z the number of atoms per cell. The coefficients of $M(k)$ are periodic functions of k , with the periodicity of the reciprocal lattice.

The normalized density of eigenvalues ν^2 associated with the matrix M can be defined by

$$G(\nu^2) = \langle \delta(\nu^2 - M) \rangle \quad (1)$$

where the average of a matrix, $\langle \rangle$, is the trace of that matrix divided by its dimensionality. Using the uniform spacing of the propagation constant over one Brillouin zone, eq. (1) can be reduced to

$$G(\nu^2) = \int \langle (\nu^2 - M(k)) \rangle dk / \int dk \quad (2)$$

where

$$\langle (\nu^2 - M(k)) \rangle = \text{trace} \int (\nu^2 - M(k)) / \ell Z = (\ell Z)^{-1} \sum_{n=1}^{\ell Z} \delta(\nu^2 - \nu_n^2(k))$$

The relation between the frequency distribution function $g(\nu)$ and the distribution function of the squares of the frequency $G(\nu^2)$, is:

$$g(\nu) = 2\nu G(\nu^2) \quad (3)$$

Since the integral in (2) cannot be evaluated explicitly except for a few special cases^{1,2,3} many approximate methods⁴ have been used

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- ¹ E. W. Montroll, J. Chem. Phys. 15, 575 (1947).
 - ² W. A. Bowers and H. B. Rosenstock, J. Chem. Phys. 18, 1056 (1950).
 - ³ G. F. Newell, J. Chem. Phys. 21, 1877 (1953).
H. B. Rosenstock, J. Chem. Phys. 21, 2064 (1953).
H. B. Rosenstock and Gordon F. Newell 21, 1607 (1953).
H. B. Rosenstock and H. M. Rosenstock 21, 1608 (1953).
 - ⁴ A discussion of some of the methods together with references is given by A. C. Menzies, Repts. Prog. Phys. 16, 83 (1953).

to study $g(\nu)$. Blackman⁵ approximates $g(\nu)$ by calculating the frequencies at a large number of points in the Brillouin zone and finding their distribution. This requires a great deal of labor, which has to be repeated for every set of force constants. Houston⁶ finds the distribution along special lines in reciprocal space and interpolates for the rest of the Brillouin zone. This method introduces some spurious singularities in $g(\nu)$.⁷ We shall discuss Montroll's^{8,9} moment method in some detail as it forms part of the basis of our work.

Using (1) the n th moment of $G(x)$ is given by

$$m_n = \int x^n G(x) dx = \langle M^n \rangle \quad (4)$$

or taking explicitly the average over the propagation constant¹⁰ as in (2)

$$m_n = \int \langle M^n(k) \rangle dk / \int dk \quad (5)$$

Montroll was the first to recognize that the moments, m_n are easily computed from traces at powers of $M(k)$ via (5). He showed that the first N moments can be used to construct the best approximate distribution function in the sense of least squares, if the approximating function can be expressed in a power series or a polynomial series contain N terms.¹¹ For these two choices of expansion, the least squares criterion

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- ⁵ M. Blackman, Repts. Prog. Phys. 8, 11 (1941).
 - ⁶ W. V. Houston, Rev. Mod. Phys. 20, 161 (1948).
 - ⁷ T. Nakamura, Prog. Theor. Phys. 5, 213 (1950).
 - ⁸ E. W. Montroll, J. Chem. Phys. 10, 218 (1942).
 - ⁹ E. W. Montroll, J. Chem. Phys. 11, 481 (1943).
 - ¹⁰ T. H. Walnut, J. Chem. Phys. 22, 692 (1954), calculates the moments directly from M .
 - ¹¹ The use of polynomials as expansion functions will always lead to the same result as the use of a power series, for the same number of terms. The use of orthogonal polynomials greatly simplifies the arithmetic.

is equivalent to the requirement that the first N moments of the approximating and exact distribution agree.¹² The rate of convergence of such an expansion is determined by the smoothness of the exact function. Hence if $G(x)$ is smooth, the use of even a few moments could be expected to give a good approximation. However, it was found by Montroll¹ when he did an exact calculation for a two-dimensional square lattice, with nearest and next nearest neighbor interactions, that the frequency distribution function was not smooth but contained two logarithmic singularities. Smollett¹³ extended Montroll's result to the case of a two-dimensional ionic lattice, taking into account the long range Coulomb forces between ions. Similar results were obtained by Bowers and Rosenstock² for the frequency distribution of vibrations perpendicular to the plane of the lattice. Van Hove¹⁴ subsequently showed that these singularities, far from being accidental, are a necessary consequence of the periodic structure of the lattice. They occur at the critical points of $\chi_n(k) = \chi_n^2(k)$, points at which $|\text{grad } \chi_n(k)| = 0$. In two dimensions $G(x)$ will have at least one logarithmic singularity. In three dimensions there will be at least three critical points x_c , where while $G(x)$ remains continuous, its derivative $G'(x_c)$ has an inverse square root singularity.

¹² If an expansion is made in functions other than polynomials, e.g. a Fourier series, the method of equating moments and the least squares method are distinct for finite N . While knowledge of the first N moments determines a Fourier expansion to N terms, if the moment conditions are applied, this knowledge is insufficient to lead to a least squares solution.

¹³ M. Smollett, J. Chem. Phys. 15, 575 (1947).

¹⁴ L. Van Hove, Phys. Rev. 89, 1189 (1953).

¶ These considerations show that an approximating function $G_a(x)$ which is smooth, such as a linear combination of polynomials which has the correct first N moments, would converge slowly to $G(x)$ with increasing N . Hence a large number of moments, and a corresponding large amount of labor, would be needed to get a good fit to $G(x)$.

We decided, therefore, to take explicitly into account the analytic nature of $G(x)$ in constructing $G_a(x)$. $G_a(x)$ is represented by an expression with the correct behavior at the singularities and at the end points. The behavior between singular points is adjusted smoothly by leaving n -undetermined parameters to be adjusted by the moments.¹⁵ In this way the moments are used for approximating a smooth function so that good agreement to $G(x)$ might be expected, even when, only a small number of moments are used. Essentially the same procedure was suggested independently by Rosenstock,¹⁶ who applied it to the body-centered and face-centered cubic lattices. See Section III for further discussion of Rosenstock's results.

II. Two Dimensional Square Lattice

As a test, this procedure, which we call the moment-singularity method was applied to a two-dimensional monatomic square lattice with neighbor and next neighbor interactions. The distribution function obtained by this procedure will be compared: (a) with the exact distribution calculated by Montroll,¹ (b) with the distribution obtained by the unmodified method of moments.

For this lattice $M(k)$ is a two by two matrix¹⁷

$$M(k) = 1/2 \begin{pmatrix} (t-1)C_1 - tC_1C_2 + 1 & tS_1S_2 \\ tS_1S_2 & 1 + (t-1)C_2 - tC_1C_2 \end{pmatrix} \quad (c)$$

where $C_i = \cos k_i$, $S_i = \sin k_i$, $i = 1, 2$, $t = (1 + \alpha/2\gamma)^{-1}$. α and γ

¹⁵ The fitting between singular points may also be done by any other method, e.g. Blackman's which yields sufficiently accurate results away from the singularities.

¹⁶ H. B. Rosenstock, On Counting Lattice Frequencies, Bull. Amer. Phys. Soc., Vol. 29, No. 4, I 10.

¹⁷ For comparison with notation in reference 1: $k_1 \rightarrow \phi_1$, $x \rightarrow f^2$, $G(x) \rightarrow (2f)^{-1} \chi(f)$. $\chi(f)$ we measure the frequency in such units that the largest frequency $\chi_L = 1$.

are neighbor and next neighbor forces respectively. The two roots of the secular equation are

$$x_{\pm} = 1/4 \left\{ 2t(1-C_1C_2) + (1-t)(2-C_1-C_2) \pm \sqrt{4t^2 S_1^2 S_2^2 + (1-t)^2 (C_1-C_2)^2} \right\}^{1/2} \quad (7)$$

The nth moment of $G(x)$ can be found from (5) by integrating the trace $M^n(k)$ over the Brillouin zone, $-\pi < k_1, k_2 \leq \pi$, and dividing the result by $2(2\pi)^2$. Thus, the first moment m_1 , is

$$m_1 = (8\pi^2)^{-1} \int_{-\pi}^{\pi} dk_1 \int_{-\pi}^{\pi} dk_2 \left\{ \frac{2-(1-t)(\cos k_1 + \cos k_2) - t \cos k_1 \cos k_2}{2} \right\} = 1/2 \quad (8)$$

Montroll has calculated the first six even moments μ_{2n} of $\mathcal{G}(\nu)$, in a slightly different way. These are related to the m_n by

$$\mu_{2n} = \int_0^1 \nu^{2n} \mathcal{G}(\nu) d\nu = \int_0^1 x^{2n} G(x) dx = m_n \quad (9)$$

and their values are

$$m_0 = 1, m_1 = 1/2, m_2 = 1/4 + \frac{(1-t)^2 + t^2}{8} \quad (10)$$

$$m_3 = 1/8 + 3 \left[(1-t)^2 + t^2 \right] / 16$$

$$m_4 = 1/16 + (3/16) \left[(1-t)^2 + t^2 \right] + (3/128) \left[(1-t)^2 + t^2 \right]^2 + (5/128) \left[(1-t)^2 + t^2 \right]$$

$$m_5 = 1/1024 (462 - 980t + 960t^2 - 265t^3 + 80t^4 - 5t^5)$$

An explicit form for all higher moments was found by T. H. Walnut.¹⁰

Due to the symmetry of the problem it is sufficient to work in one quadrant of the Brillouin zone, $0 \leq k_1, k_2 \leq \pi$. In this quadrant the critical points of $x_{\pm}(k)$ are at, $(k_1, k_2) = (0,0), (0,\pi), (\pi,0), (\pi,\pi)$ when $0 < t \leq 1/5$.

The values of x , in the two branches $x_{\pm}(k)$, at these critical points are $(C, 1, 1, 1-t)$ and $(0, t, t, 1-t)$ respectively. For t greater than $1/5$ but less than $1/2$,¹⁸ the x_+ branch has one additional critical point at $[\cos^{-1}(t-1/4t), \cos^{-1}(t-1/4t)]$ and the value of $x_+(k)$ at that point is $(1+3t)^2/(16t)$.

¹⁸ We shall assume as did Montroll in ref. 1 that $t < 1/2$.

From the behavior of $x_{\pm}(k)$ in the neighborhood of the critical points in reciprocal space it is possible to deduce the form of $G(x)$ near the critical values of x ,¹⁹ including the value of $G(0)$ and $G(1)$.

If the singular part of $G(x)$ is called $F(x)$, then it can be shown that

$$F(x) = \begin{cases} B_1 \ell_n |x-t| + B_2 \ell_n |x-(1-t)| + B_3 H(x-(1-t)), & 0 < t < 1/5 \\ C_1 \ell_n |x-t| + C_2 \ell_n |x-(1+3t)^2/16t| + C_3 H(x-(1-t)), & 1/5 < t < 1/2 \end{cases} \quad (12a)$$

where H is the Heaviside unit function $H(y) = 0, y < 0, H(y) = 1, \text{ for } y > 0$, and

$$\begin{aligned} B_1 &= -[t(1-2t)]^{-1/2}/\pi^2, B_2 = -(1-3t)[t(1-t)(1-2t)(1-5t)]^{-1/2}/\pi^2 \\ B_3 &= -(2/(t\pi^2)) \int_0^{\pi/2} d\theta \{ (1-3t)/t + (1+k^2 \sin^2 \theta)^{1/2} \}^{-1}, k^2 = (3t^2 - 2t + 1)/t^2 \\ C_1 &= -[t(1-2t)]^{-1/2}/\pi^2; C_2 = [16t/\pi^2] (1-t)^{-1} [3(5t-1)(3t+1)]^{-1/2} \\ C_3 &= (1-3t) [t(1-t)(1-2t)(5t-1)]^{-1/2} \end{aligned} \quad (12b)$$

The value of $G(x)$ at 0 and 1 is

$$G(0) = (1+t) [t(1-2t-3t^2)]^{-1/2}/\pi; G(1) = t^{-1/2}/\pi \quad (12c)$$

In order to apply the moment-singularity method we subtracted $F(x)$ from $G(x)$ and approximated the remainder by a linear combination of n Legendre polynomials. The first $n-2$ coefficients of the expansion are determined by the requirement that first $n-2$ moments of the approximate function agree with the exact ones. The remaining two coefficients are then determined by fixing the end points. Hence, when N moments are known, the approximate distribution function $G_a^{(N)}(x)$ is:

$$G_a^{(N)}(x) = F(x) + \sum_{k=0}^{N+1} A_k^{(N)} P_k(2x-1), \quad 0 \leq x \leq 1 \quad (13a)$$

where

$$A_k^{(N)} = (2k+1) \left[\int_0^1 P_k(2x-1) G(x) dx - \int_0^1 P_k(2x-1) F(x) dx \right] \quad (13b)$$

¹⁹

For a discussion see Section III of this paper, also Van Hove, reference (14) sections II and III. For comparison see Montroll, reference 1.

for $k \leq N-1$

and

$$A_N^{(N)} + A_{N+1}^{(N)} = G(1) - F(1) - \sum_{i=0}^{N-1} A_i^{(N)} \quad (13)$$

$$(-1)^N A_N^{(N)} + (-1)^{N+1} A_{N+1}^{(N)} = G(0) - F(0) - \sum_{i=0}^{N-1} (-1)^i A_i^{(N)}$$

Note that $A_k^{(N)}$ is independent of N , the order of the approximation.

$$A_k^{(N)} = A_k^{(CC)}, \text{ for } k \leq N-1.$$

When the unmodified method of moments is used to approximate $G(x)$, the approximation function, when N moments are known, $G_m^{(N)}(x)$ is

$$G_m^{(N)}(x) = \sum_{k=0}^{N-1} B_k^{(N)} P_k(2x-1) \quad (14)$$

the $B_k^{(N)}$ are again given by eq. (13b) with $F(x)$ set equal to zero.

A numerical evaluation of the $A_k^{(N)}$ and the $B_k^{(N)}$ was made for $t = 1/3$. This is the value of t for which Montroll^{1,17} evaluated $G(x)$ in a closed form, so that comparisons can be made. This will indicate how useful the moment-singularity method is.

For $t = 1/3$, eqs. (12) and (13) yield

$$G_a^N(x) = -3/\pi^2 \ln |x-1/3| - 4/\pi^2 \ln |x-3/4| + \sum_{k=0}^{N+1} A_k^{(N)} P_k(2x-1) \quad (15)$$

$$G(0) = 2/\pi; G(1) = (3)^{1/2}/\pi$$

The values of the $A_k^{(N)}$ in different orders of approximation $N \leq 6$ are summarized in table 1. The values of the $B_k^{(N)}$, which are independent of N , are $B_0 = 1$, $B_1 = 0$, $B_2 = -.4167$, $B_3 = 0$, $B_4 = -.2269$, $B_5 = -.2290$ (16) Comparisons between $G(x)$, $G_a^N(x)$, $G_m^N(x)$ are presented in tables 2, 3, and 4.

III. Conclusion

As can be seen from Table 4 use of the non-singular method of moments results in a great increase in the accuracy of the approximation over the usual method of moments for the same number of moments.

The additional work required to find $F(x)$ was small, for $t = 1/3$.

For the particular value of t , $x^\pm(k)$ can be expanded in a Taylor series near the critical points, and the behavior of $G(x)$ near the critical points can be read off directly from Van Hove. For other values of t , the critical points $(0, 0)$, (π, π) are of the type which Van Hove calls generalized critical points. The behavior of $x^\pm(k)$ near these points is

$$x(k) = x_c + |\xi|^2 \psi(\xi/|\xi|) + O(|\xi|^3), \quad \xi = \vec{k} - \vec{k}_c$$

To find the form of $G(x)$ near these points the integral in (2) has to be evaluated, neglecting terms of $O(|\xi|^3)$. Transforming to polar coordinates in reciprocal space the integration over the radial variable, can be done immediately by means of the delta function. This leaves an integration over the angle variable whose evaluation may be quite difficult, if it cannot be found in the tables. The integral representing B_3 in eq. (12b) is one such case.

However even when the exact form of the singular part of $G(x)$, such as the coefficient multiplying the logarithm terms in two dimensions, or the square root terms in three dimensions, is not known, convergence would be improved greatly if the approximating function contained terms of the correct singular form. The coefficients of these terms could then be determined by the moments. Thus, for the case considered in this paper, it would have been preferable if the approximating function $G_a(x)$ consisted of polynomials multiplying the logarithmic terms whose value at the critical points is the correct one. This would have taken account of terms in the distribution function of the form $(x-x_c) \ln |x-x_c|$. A smaller number of moments might then have been used to obtain the same accuracy. This was not done because of the extra

work involved in solving simultaneous equations for the coefficients of the multiplying polynomials since no orthogonal set would be available. In general the amount of work required to find more moments has to be balanced against the work involved in solving simultaneous equations.

There is a theorem of Feynman²⁰ which is useful in finding some critical points without diagonalizing $M(k)$. This theorem states, if $A(\alpha) \psi(\alpha) = \lambda(\alpha) \psi(\alpha)$, $A(\alpha)$ is hermitian.

Then

$$\frac{\partial \lambda(\alpha)}{\partial \alpha} = \left(\psi(\alpha), \frac{\partial A(\alpha)}{\partial \alpha} \psi(\alpha) \right)$$

Thus $|\text{grad } \nu^2(k)| = 0$ whenever $|\text{grad } M(k)| = 0$. This can happen only at those k , where all the branches have critical points simultaneously.

A great difficulty, in many cases, in finding the analytic form of the distribution, is the location of all the critical points. Van Hove's arguments predict only the minimum number of critical points but give no upperbound. It is generally easy to find those which are at symmetry points of the Brillouin zone. In most cases investigated thus far^{1,2,3} these are the only critical points. This might be due to the assumption made in these cases that the forces are short range. The number of critical points might be expected to increase with the range of the interaction and their location is then more likely to be at non-symmetry points of the zone. Rosenstock¹⁶ has devised a method for examining the presence of critical points inside the zone from the behavior of $\nu^2(k)$ on the boundary.

For three dimensions, with short range forces, when the critical points are at the symmetry points the matrix $M(k)$ is easy to diagonalize

²⁰ R. P. Feynman, Phys. Rev. 56, 340 (1939).

at those points to find the critical frequencies. To find the exact form of the distribution near the critical points x_c , degenerate perturbation theory has to be used to find the eigenvalues in the vicinity of k_c . However, since in three dimensions $G(x)$ does not become infinite at x_c it might be sufficient to use the right form of $G(x)$ near x_c , the coefficients of the singular part would then be determined by the moment as mentioned above. This would agree with the results of Rosenstock.¹⁶

Table 1

Value of the coefficients of A_k^N of the Legendre polynomials
in the expansion of the non-singular part of distribution function.

$N \backslash k$	A_0^N	A_1^N	A_2^N	A_3^N	A_4^N	A_5^N	A_6^N	A_7^N
$G^{(1)}$	-.1306	-.1599	.1568					
$G^{(2)}$	-.1306	-.2620	.1568	.1021				
$G^{(3)}$	-.1306	-.2620	.1409	.1021	.0159			
$G^{(4)}$	-.1306	-.2620	.1409	.1383	.0159	-.0368		
$G^{(5)}$	-.1306	-.2620	.1409	.1388	.0536	-.0368	-.0378	
$G^{(6)}$	-.1306	-.2620	.1409	.1388	.0536	-.0429	-.0378	.0061

Table 2

Convergence of the moment-singularity approximation $G_a^{(N)}(\sqrt{2})$ to the exact distribution $G(\sqrt{2})$ with increasing N . (N is the number of moments used.)

[illegible]

Table 3

Convergence of the unmodified moment approximation $G_m^{(N)}(\psi^2)$, to the exact distribution $G(\psi^2)$ with increasing N . (N is the number of moments used.)

ψ^2	$G_m^{(1)}(\psi^2)$	$G_m^{(2)}(\psi^2)$	$G_m^{(3)}(\psi^2)$	$G_m^{(4)}(\psi^2)$	$G_m^{(5)}(\psi^2)$	$G_m^{(6)}(\psi^2)$	$G(\psi^2)$
0	1	.583		.332	.103	.637	
.05	1	.702		.655	.646	.683	
.1	1	.808		.861	.770	.740	
.3	1	1.11		1.13	1.20	1.34	
1/3	1	1.14		1.14	1.21	∞	
.4	1	1.18		1.13	1.20	1.17	
.5	1	1.21		1.12	1.12	.951	
.6	1	1.18		1.13	1.06	.908	
.7	1	1.11		1.13	1.07	1.14	
.75	1	1.05		1.12	1.09	∞	
.9	1	.808		.861	.953	.733	
.95	1	.702		.655	.664	.631	
1	1	.583		.332	.561	.551	

Table 4

Comparison of the unmodified moment approximation $G_n^{(6)}(y^2)$, the moment singularity approximation $G_a^{(6)}(y^2)$, and the exact distribution $G(y^2)$, when six moments are used.

x	$G_n^{(6)}(x)$	$G_a^{(6)}(x)$	$G(x)$
0	.103	.637	.637
.05	.646	.639	.683
.1	.770	.739	.740
.3	1.20	1.35	1.34
1/3	1.21	∞	∞
.4	1.20	1.18	1.17
.5	1.12	.937	.951
.6	1.06	.887	.908
.7	1.07	1.16	1.14
.75	1.09	∞	∞
.9	.953	.695	.733
.95	.664	.619	.631
1	.561	.551	.551

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